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OPTICAL GENERATION AND DETECTION OF NEAR-ZONE-BOUNDARY PHONONS IN RUBY

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A new scheme for phonon spectroscopy near the zone boundary using luminescent centers is discussed. Experimental results in optically pumped ruby are presented.

Optically pumped luminescent centers are now widely used as generators and detectors of high-frequency phonons in crystals. Monochromatic phonon generation and detection taking advantage of a direct phonon associated transition at the luminescent center have for the first time been demonstrated in ruby by observing the R_1 and R_2 fluorescent intensities as a function of the pumping power and a magnetic field[1]. The optical techniques have made high-frequency phonons accessible to experiment, thus allowing important new information to be obtained. These schemes have however not yet been extended to probing acoustic phonons close to the zone boundary. It is this part of the phonon spectrum which has recently drawn much attention owing to the observation of surprisingly long life-times[2,3], and the theoretical result that at low temperatures TA phonons near the zone boundary are stable against anharmonic decay[4]. The experiments are however too few to be conclusive. Here we present a new optical scheme of phonon spectroscopy which is inherently selective for near-zone-boundary phonons. For the detection, the scheme employs, instead of a direct phonon-associated transition, second-order Raman transitions connecting two luminescent states. As an example we consider the case of diluted ruby.

The generation of the phonons takes place at the Cr^{3+} impurity center through the fast nonradiative decay from the broadbands to the metastable state ^2E following optical excitation. Since conservation of $\vec{k}$ is not implied, the acoustic phonons are preferentially generated near the zone boundary by virtue of the high density of states there. When sufficiently long-lived, these phonons can be driven into a bottleneck situation by optical pumping[1,5]. Such a nonequilibrium population is reflected in a redistribution of the luminescent R_1 and R_2 intensities, emerging from $\bar{E}(^2E)$ and $2\bar{A}(^2E)$, respectively, due to two-phonon Raman processes, essentially populating $2\bar{A}$ out of $\bar{E}$. Accordingly the enhancement of the fluorescent intensity, ΔR_2, is a direct gauge for the average occupation number of near-zone-boundary phonons, p_{ZB}, through

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$$\Delta R_2 = \alpha(p_{ZB}/T_{Ram})T_{eff}R_1. \quad (1)$$

Here, p_{ZB}/T_{Ram} is the effective Raman rate connecting $\bar{E}$ and $2\bar{A}$, T_{eff} is the effective relaxation time of $2\bar{A}$, R_1 is the fluorescent intensity of $\bar{E}$, and α is a known constant of order unity. For ruby T_{Ram} is calculated to be of order 10^{-11} s [6], while T_{eff}, dependent on the bottlenecking[1], typically is of order 0.1 - 1 μs. The key to p_{ZB} therefore is to arrange the experiment such that ΔR_2, R_1, and T_{eff} are determined in a single measurement. This is feasible with modulated pumping on a time scale short compared to the radiative lifetime (3.7 ms in the absence of trapping), but long relative to T_{eff}. During the "light-on" period R_1 and ΔR_2 are measured, while T_{eff} is extracted from the uniexponential decay of ΔR_2 after switching off the optical pumping.

In the experiments, optical generation and detection of near-zone-boundary phonons in ruby (130, 700, and 2500 ppm) were achieved at 1.5 K upon pumping with a 3W argon laser, operating at 514 nm or 458 nm. The former laser line has a larger *ground-state* absorption cross-section (by a factor of 4), whereas the latter has a stronger *excited-state* absorption from ^2E (by a factor of 14; $\vec{E}||\vec{c}_3$)[7]. The laser was switched with an acousto-optic modulator. The luminescent lines were selected by a double monochromator followed by photon-counting equipment. Time resolution was obtained by a time-to-amplitude converter.

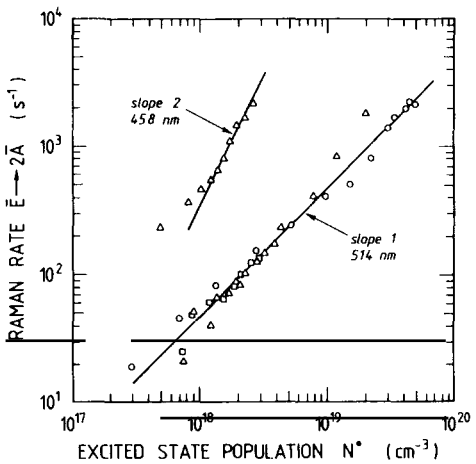

FIGURE 1

Raman rate connecting $\bar{E}$ and $2\bar{A}$ induced by optically produced zone-boundary phonons vs. the excited-state population N^* for 130 (▫), 700 (▵) and 2500 ppm (○) ruby at 1.5 K. Pumping is done with the 514 nm line (lower curve) or 458 nm line (upper curve) of an argon laser.

Figure 1 shows the effective Raman rate p_{ZB}/T_{Ram}, as extracted from ΔR_2, R_1, and T_{eff} according to Eq.(1), vs. the excited state population N^*. The horizontal scale is calibrated on the basis of the saturation behavior of R_1 vs. the pump power. Adjustment of the vertical scale [α in Eq.(1)] is performed for the various runs to account for the effects of trapping of the luminescence. A linear dependence of the Raman rate is found up to 10^3 s^{-1} when pumping at 514 nm. This corresponds to a production of near-zone-boundary phonons proportional to the optical pump rate up to occupation numbers of 10^{-8}, which is equivalent to a nonequilibrium mode temperature of about 15 K. Upon pumping at 458 nm well below saturation of R_1, however, a quadratic dependence appears, suggestive for excited-state absorption from $\bar{E}$ followed by fast nonradiative decay back to $\bar{E}$, and, as a consequence, an increased phonon generation near the zone boundary. These findings are quantitatively consistent with earlier results[8], if one assumes the effective phonon lifetime around the zone boundary to be of order 0.1 - 1 μs, and roughly proportional to the characteristic size of the excited zone. This assumption is further corroborated by experiments, not presented here, to the effect that the phonon lifetime is linearly dependent on the zone dimensions in the range 50 - 500 μm.

In conclusion, the present experiments suggest for zone-boundary phonons (i) efficient production by optical pumping of the broad bands, (ii) slow thermalization, and (iii) lifetimes of the order of one microsecond determined by ballistic flight out of the excited zone.

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